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Nuclear Instruments and Methods in Physics Research B 249 (2006) 451–453

NIM B
Beam Interactions with Materials & Atoms

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Combined RBS and TEM characterization of nano-SiGe layers embedded in SiO_2

A. Kling [a,b,*], M.I. Ortiz [c], J. Sangrador [c], A. Rodríguez [c], T. Rodríguez [c], C. Ballesteros [d], J.C. Soares [b]

[a] *Instituto Tecnológico e Nuclear, Estrada Nacional no. 10, 2686-953 Sacavém, Portugal*
[b] *Centro de Física Nuclear da Universidade de Lisboa, 1649-003 Lisboa, Portugal*
[c] *Departamento de Tecnología Electrónica, E.T.S.I. de Telecomunicación, Universidad Politécnica de Madrid, 28040 Madrid, Spain*
[d] *Departamento de Física, E.P.S., Universidad Carlos III, 28911 Leganés (Madrid), Spain*

Available online 24 May 2006

Abstract

Grazing incidence RBS has been tested as a technique to detect and characterize SiGe nanoparticles embedded in a SiO_2 matrix. Suitable structures were deposited by low pressure chemical vapour deposition and characterized by TEM and RBS. The layers containing nanoparticles have been modelled by stacks of sublayers consisting of SiGeO layers with compositions calculated according to presumed shapes, sizes, Si/Ge ratios and particle area densities and used as input for RUMP. The nanoparticle parameters obtained by fitting the experimental RBS spectra agree well with the findings by TEM. This demonstrates that RBS is a useful and fast technique to characterize this kind of structures.

PACS: 68.65.Jk; 82.80.Yc; 78.30.Am

Keywords: Rutherford backscattering spectrometry; SiGe nanoparticles; Transmission electron microscopy; Chemical vapour deposition

1. Introduction

Ge or Si nanoparticles embedded in a SiO_2 matrix are used for nanomemories [1] and optoelectronic applications [2]. A promising approach for their fabrication is the use of a conventional low pressure chemical vapour deposition (LPCVD) system, since it allows to form SiGe nanoparticles of any composition and to process many wafers simultaneously [3].

Transmission electron microscopy (TEM) permits to characterize the shape, crystallography and size of the nanoparticles. Also rough estimates of their composition can be obtained by energy dispersive X-ray emission (EDX). Since the technique is very time consuming it would be interesting to investigate the existence of nanoparticles and their characteristics by Rutherford backscattering spectrometry (RBS).

In this paper we compare the results obtained by grazing incidence RBS and TEM studies of samples containing SiGe nanoparticles embedded in SiO_2. For the analysis of the RBS spectra we model the nanoparticle containing region by a stack of SiGeO layers with different atomic fractions.

2. Experimental details

SiO_2/SiGe/SiO_2 structures have been deposited onto Si wafers, in a continuous process, using a commercial LPCVD reactor. Germane (GeH_4) and disilane (Si_2H_6) were used as precursor gases for the SiGe deposition, while the SiO_2 was deposited from Si_2H_6 and O_2. The GeH_4 to

* Corresponding author. Address: Instituto Tecnológico e Nuclear, Estrada Nacional no. 10, 2686-953 Sacavém, Portugal. Tel.: +351 21 994 6154; fax: +351 21 994 1039.
E-mail address: akling@itn.mctes.pt (A. Kling).

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doi:10.1016/j.nimb.2006.03.029

Si_2H_6 flow ratio was kept at 0.82. The temperature was 390 °C and the pressure was 50 mTorr resulting in a reduced deposition rate (around 5 nm/h range). All SiO_2 layers have a nominal thickness of 20 nm. More details on the system and on the deposition process can be found elsewhere [4]. Four runs have been deposited for the purposes of this work. The deposition times were 60, 25, 12.5 and 6 min (samples A, B, C and D, respectively).

Grazing incidence RBS studies were done using a 2 MeV $^4He^+$ ion beam impinging on the target tilted to 78°. The Si surface barrier detector was located at 160° with respect to the incident beam in Cornell geometry.

SiGe cylindrical and spherical nanoparticles embedded in SiO_2, were modelled as a single or a stack of SiGeO layers, respectively. For the simulation of cylindrical nanoparticles in a SiO_2 matrix the atomic composition (Si, Ge, O) of a layer with the height of the cylinder was calculated taking into account their diameter, Ge-fraction and the particle area density. For the case of spherical nanoparticles a layer with a thickness equal to the diameter of the sphere was divided into seven sublayers containing disks with adequate dimensions. The atomic composition of these sublayers was calculated as in the previous case. The layer structures were used as input for simulations with the RUMP code [5].

Cross-sectional specimens suitable for high-resolution transmission electron microscopy (HRTEM) were prepared by standard procedures. TEM images were obtained using a Philips Tecnai 20F FEG analytical microscope operating at 200 keV, equipped with a dark field high angle annular detector (HAAD) for Z-contrast analysis and EDX.

3. Results and discussion

The study of the layer systems by TEM indicates that only the SiGe layer of sample A is continuous and 4–6 nm thick. Fig. 1(a) and (b) show TEM images of samples B, C and Fig. 1(c) a Z-contrast STEM image of sample D (TEM image of this sample shows only a very weak contrast). A uniform distribution of spherical nanoparticles, located in the same plane, is found. This means that the deposition process was stopped before the coalescence of growth nuclei could occur. EDX spectra indicate that the nanoparticles are SiGe. The size of the nanoparticles was in the ranges of 3.2–4.6, 3–4 and <2.5 nm for samples B, C and D, respectively. A rough estimate of density of nanoparticles is in the order of 2×10^{12} cm^{-2} (see Table 1).

In order to test the feasibility of our approach to detect discontinuous layers by RBS and deduce useful information about the characteristics of nanoparticles various comparative simulations have been performed for particles with spherical and cylindrical shape and continuous layers of Ge. As an example, Fig. 2(a) shows the results for a continuous $Si_{0.7}Ge_{0.3}$ layer with a nominal thickness of 0.063 nm, spherical nanoparticles of 5 nm diameter and cylindrical ones with 5 nm diameter and 3.2 nm height (identical amount of SiGe in the layer and number of particles), all embedded in two layers of SiO_2. While for both types of nanoparticles the area under the Ge-peak is virtually the same a shift of the Ge-peak centre to lower energies is observed. The smaller shift observed for cylindrical particles is mainly due to the lower thickness of the layer. Fig. 2(b) demonstrates the influence of the particle diameter in the case of spherical nanoparticles. Besides the expected increase of the Ge-peak area with increasing particle size also a shift of the peak centre to lower energies is observed.

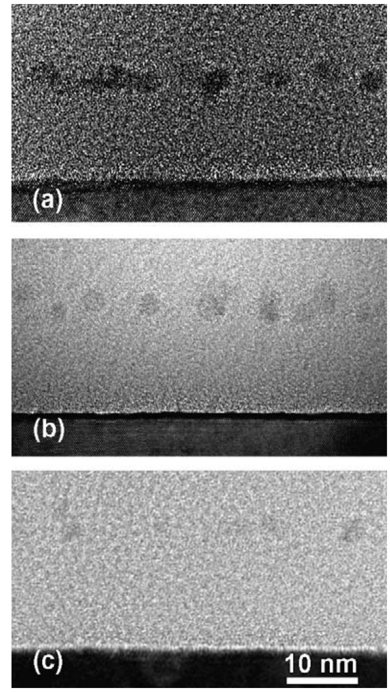

Fig. 1. Cross-sectional HREM image of: (a) sample B, (b) sample C. (c) Z-contrast STEM of sample D. Underfocus/overfocus conditions were selected to increase the visibility contrast of the nanoparticles.

Table 1
Parameters of the best RBS fits for spherical nanoclusters and embedding SiO_2 layers of all four samples

Sample	Equivalent thickness (nm)	Spherical nanoclusters		TEM	
		Diameter (nm)	Particle density (cm^{-2})	Diameter (nm)	Particle density (cm^{-2})
A	4.6	–	–	–	–
B	1.1	4.6	2.1×10^{12}	3.2–4.6	$<2.8 \times 10^{12}$
C	0.40	3.3	1.8×10^{12}	3–4	$<1.3 \times 10^{12}$
D	0.20	2.5	1.8×10^{12}	<2.5	–

The applicability in actual discontinuous layer structures was tested for the same samples as characterized by TEM. Fig. 3 shows the RBS spectra obtained for the samples B–D together with the best fits assuming spherical nanoparticles with the composition $Si_{0.6}Ge_{0.4}$. The sizes and particle densities obtained by RBS are summarized in Table 1. It has to be noted that also assuming cylindrical

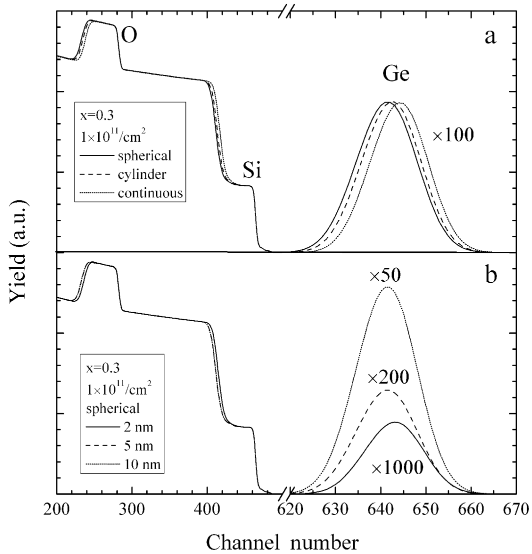

Fig. 2. Comparison of simulated RBS spectra assuming the existence (a) of spherical and cylindrical SiGe nanoparticles or a continuous SiGe layer embedded in SiO_2 and (b) of spherical SiGe nanoparticles with different diameters.

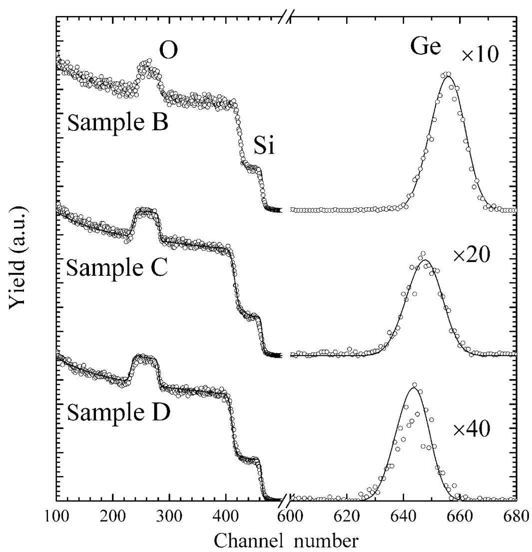

Fig. 3. Comparison of the experimental spectra (circles) for samples B–D with the best fit obtained for each one (solid line) using spherical nanoparticles.

particles good fits are obtained if the diameter was equal to that of the best value for spheres and their height roughly equal to their diameter, i.e. when they approximate best a spherical shape. Therefore a spherical shape seems to be the most appropriate solution.

Although the sample spectra could not be fitted assuming a continuous SiGe layers sandwiched by to SiO_2 layers, except for sample A which is continuous, "equivalent" thickness for the SiGe layers of 4.6 nm (A), very close to the value obtained by TEM, 1.1 nm (B), 0.40 nm (C) and 0.20 nm (D) can be extracted from the Ge-peak assuming the composition of $Si_{0.4}Ge_{0.6}$. While the thickness of the SiO_2 layer deposited on the Si substrate shows only slight variations (between 15 and 18 nm) for the layer on top of the SiGe a increase from 10 nm for the thickest to 29 nm for thinnest SiGe layer is observed which is related to the incubation time for the deposition of SiO_2 on SiGe.

4. Summary

A model for the simulation RBS spectra of nanoparticles embedded in a dielectric matrix has been developed. In order to check its validity, SiGe nanoparticles of different sizes, embedded in a SiO_2 matrix have been deposited by LPCVD and characterized by TEM. It was found that the analysis of RBS spectra obtained under grazing incidence can yield indications on the presence of nanoparticles, their size and density. Therefore this technique enables a rapid screening of samples for the existence of nanoparticles and their characteristics in order to select only the most interesting ones for more time consuming studies, e.g. TEM.

Acknowledgements

This work was funded by Spanish Government through CICYT Project MAT2004-04580-C02-01 and a common grant by CRUP (Portugal) and MCT (Spain) in the framework of the Acções Integradas/Acciones Integradas program.

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